

## Low-Energy Physics of Hole Doped $\text{Y}_2\text{BaNiO}_5$

In a recent Letter [1], a low-energy (LE) reduction from a multiband Hamiltonian for hole doped  $\text{Y}_{2-x}\text{Ca}_x\text{BaNiO}_5$  to a one-band model  $H_1$  was discussed. Calculating numerically the dynamical structure factor  $S(q, \omega)$  of  $H_1$ , an explanation of the observed peaks below the Haldane gap [2] was presented. Unfortunately we find that:

(1) *The starting multiband Hamiltonian is incorrect.*— It is important to treat accurately the highest energy scales before integrating them out. These energies are the correlations within the Ni 3D shell. Including only the  $e_g$  orbitals, they take the form

$$H_d = U_d \sum_{i\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow} + (U_d - J_{\text{Hund}}) \sum_{i\sigma\sigma'} n_{i1\sigma} n_{i2\sigma'} + \frac{J_{\text{Hund}}}{2} \left[ \sum_{i\sigma\sigma'} d_{i1\sigma}^\dagger d_{i2\sigma'}^\dagger d_{i1\sigma'} d_{i2\sigma} + \sum_i (d_{i1\uparrow}^\dagger d_{i1\downarrow}^\dagger d_{i2\downarrow} d_{i2\uparrow} + \text{H.c.}) \right],$$

where in terms of Slater parameters  $U_d = F_0 + 4F_2 + 36F_4$  and  $J_{\text{Hund}} = 8F_2 + 30F_4$ . From atomic data and the physics of NiO [3],  $F_0 \sim 10$  eV  $\gg F_2 \sim 0.16$  eV  $\gg F_4 \sim 0.01$  eV. Thus, the first and second terms of  $H_d$  are an order of magnitude larger than the other two. In Ref. [1], only the first and third terms are retained, breaking the  $O_h$  symmetry by  $\sim 9$  eV [4]. This lowers the ground state energy of  $\text{Ni}^{+3}$  ( $\text{Ni}^{+2}$ ) by  $\sim 18$  eV ( $\sim 9$  eV). Within  $\text{Ni}^{+2}$ , the state  $(d_{1\uparrow}^\dagger d_{1\downarrow}^\dagger - d_{2\uparrow}^\dagger d_{2\downarrow}^\dagger)|0\rangle$  is degenerate with  $(d_{1\uparrow}^\dagger d_{2\downarrow}^\dagger - d_{1\downarrow}^\dagger d_{2\uparrow}^\dagger)|0\rangle$  in the correct  $H_d$  and participates in effective O-O hopping processes of order  $t_{pd}^4$ . With the Hamiltonian Equation (1) of Ref. [1], the energy of this state with respect to the ground state is displaced from  $J_{\text{Hund}}$  to  $U_d$ . In addition, solving exactly the correct Hamiltonian in the basic  $\text{NiO}_6$  cluster, we find that neglecting O atoms which are not in the NiO chains has the effect of reducing by a factor of  $\sim 1.5$  the effective O-O hopping terms and exchange constant  $J_K$  (see below).

(2)  *$H_1$  misses a crucial term.*— An O hole can hop to the position of a nearest-neighbor (NN) Ni hole or vice versa if both holes have opposite spin, leading to an effective interaction  $J_K \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$  between the O spin  $\mathbf{S}_j$  and both NN Ni spins, where  $J_K = t_{pd1}^2 [1/\Delta + 1/(U_d - J_{\text{Hund}} - \Delta)]$  and the charge transfer energy  $\Delta \sim 6.2$  eV is defined as in Ref. [3]. Taking also  $t_{pd1}$  from Ref. [3] scaled with an  $r^{-7/2}$  law leads to  $J_K \sim 2.2$  eV. A more accurate estimate [5] gives  $J_K \sim 1.4$  eV. This antiferromagnetic interaction stabilizes the Zhang-Rice doublets (ZRD's) (see Fig. 1 of Ref. [1]). Since the O spins are not compensated in the ZRD's, they interact via  $J_K$  with their NN Ni spins, leading to a ferromagnetic interaction  $-J' \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$  between the spin  $\hat{\mathbf{S}}_j$  of a ZRD and its NN Ni spin, with  $J' = (\sqrt{2/3} - \sqrt{1/3})J_K \sim 0.3$  eV.

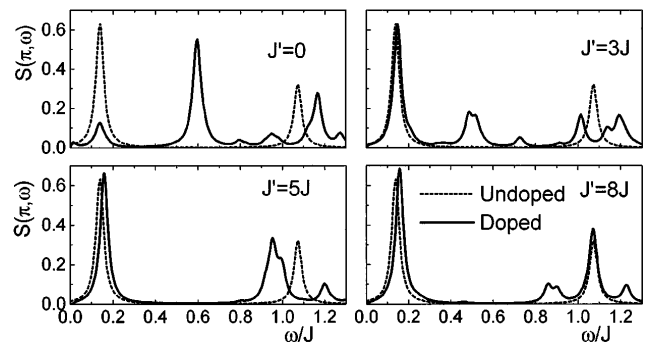


FIG. 1.  $S(\pi, \omega)$  for ten sites,  $t = 2J$  [1] and several values of  $J'$ .

$J'$  excluded in Ref. [1] is much larger than the values of  $t$  (and  $J \sim 0.023$  eV) used in Ref. [1]. The electron-phonon interaction which localizes the carriers in some NiO systems cannot modify  $J'$  or  $J_K$  by more than  $\sim 30\%$ . As shown in Fig. 1, the effect of  $J'$  on the states below the Haldane gap is dramatic. To avoid frustration effects, we used open boundary conditions (BC) [6].  $S(\pi, \omega)$  for the undoped system has a peak at low energies due to end states (which should disappear in the thermodynamic limit) and another one for  $\omega \sim J$  which identifies the Haldane peak. The additional low-energy peak for the doped system shifts first to lower energies with increasing  $J'$  and then to higher energies for  $J' \geq 3J$ . For  $J' = 8J$ ,  $S(\pi, \omega)$  for the doped system is very similar to that of the undoped one. Thus, no new LE peaks appear using  $H_1$  for realistic  $J'$  and other parameters as in Ref. [1]. However, a systematic LE reduction is able to explain the additional LE peaks [5].

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- [1] E. Dagotto *et al.*, Phys. Rev. Lett. **76**, 1731 (1996).
- [2] J.F. Di Tusa *et al.*, Phys. Rev. Lett. **73**, 1857 (1994).
- [3] J. van Elp *et al.*, Phys. Rev. B **45**, 1612 (1992).
- [4] The  $O_h$  symmetry is broken by the hopping. Crystal-field corrections to  $H_d$  should be much smaller than the cubic splitting  $10Dq$  ( $\sim 0.7$  eV in NiO [3]).
- [5] C.D. Batista, A.A. Aligia, and J. Eroles, Europhys. Lett. **43**, 71 (1998).
- [6] For  $t \sim J$ , the use of periodic BC leads to spurious peaks in  $S(q, \omega)$  below the Haldane gap due to the frustration of one bond. This frustration does not exist for  $t = 0$ . In this case low-energy peaks also exist for  $J' = 0$ , but not for realistic  $J' > J$ .